9

Soil and Sediment Monitoring

Gretchen M. Gallegos

Introduction

Soil is weathered material, mainly composed of disintegrated rock and organic material, that will sustain growing plants. Soil can contain pollutants originally released directly to the ground, to the air, or through liquid effluents. DOE guidance for environmental monitoring (U.S. Department of Energy 1991) states that soil should be sampled to determine if there is measurable long-term buildup of radionuclides in the terrestrial environment and to estimate environmental radionuclide inventories. The guidance recommends that radionuclides specific to a particular operation or facility as well as those that occur naturally should be monitored. Particulate radionuclides are of major interest in the LLNL soil monitoring program because airborne particulate releases are the most likely pathway for LLNL-induced soil contamination.

Sediments are defined, for the purposes of this chapter, as finely divided solid materials that have settled out of a liquid stream or standing water. To evaluate current conditions, LLNL samples recent sediments in storm drainage channels and the two arroyos on site. The accumulation of radioactive materials in sediment could lead to exposure of humans through ingestion of aquatic species, through sediment resuspension into drinking water supplies, or as an external radiation source (U.S. Department of Energy 1991). Note, however, that the Livermore site and Site 300 do not have habitats for aquatic species that are consumed by people, nor do they have surface drainage that directly feeds drinking water supplies. In addition, subsurface sediment sampling is conducted to support the LLNL Ground Water Protection Management Program (Chapter 8).

Since 1971, surface soil sampling in the vicinity of the Livermore site and Site 300 has been part of a continuing LLNL monitoring program designed to measure any changes in environmental levels of radioactivity and to evaluate any increase in radioactivity that might have resulted from LLNL operations. These samples have been analyzed for plutonium and gamma-emitting radionuclides, such as depleted uranium, which is used in some high-explosives tests at Site 300. The inclusion of other gamma-emitting naturally occurring nuclides (^{40}K and ^{232}Th) and the long-lived fission product ^{137}Cs provides background information and baseline data on global fallout from historical aboveground nuclear weapons testing.

Similarly, sediment samples have been collected from selected arroyos and other drainage areas at and around the Livermore site since 1988; these locations largely coincide with selected storm water sampling locations (see Chapter 7). The number of sediment sampling locations was reduced in 1994 to correspond to reductions in storm water sampling locations. In addition, in 1991, LLNL began analyzing surface soil samples for beryllium, a potentially toxic metal used at both the Livermore site and Site 300. However, analysis for beryllium was discontinued at the Livermore site in 1995, because beryllium was not ever measured above background values.

Location maps for soil and sediment sampling conducted during 1995 are provided in **Figures 9-1** through **9-3**. The locations were selected to represent background concentrations (distant locations unlikely to be affected by LLNL operations) as well as areas where there is the potential to be affected by LLNL operations. Areas with known contaminants, such as the Livermore Water Reclamation Plant (LWRP), are also sampled. In general, Site 300 soil sampling locations were established around firing tables and other areas of potential soil contamination. Arroyo and drainage channel sediment sampling locations were chosen to coincide with major Livermore site storm water drainages. All soil and sediment sampling locations have permanent location markers for reference.

Methods

Soil and sediment sampling is conducted according to written, standardized procedures (Tate et al. 1995, Appendix A). Soil samples are collected from undisturbed areas near the permanent sampling location marker. These areas generally are level, free of rocks, and are unsheltered by trees or buildings. All samples are collected from the top 5 cm of soil because surface deposition from the air is the primary pathway for potential contamination. Quality control duplicate samples are submitted with each batch of soil samples. At locations chosen for this sampling, two identical samples are collected.

Samples of recent sediment are collected annually from drainages at and around the Livermore site after the cessation of spring runoff. Sediment sampling location ASS2 was not sampled in 1996 because of problems with accessibility. For 1996, samples at the Livermore site were analyzed for radionuclides and samples for Site 300 were analyzed for radionuclides and beryllium. During 1996, additional subsurface sediment sampling supported the LLNL Ground Water Protection Management Program (Chapter 8).

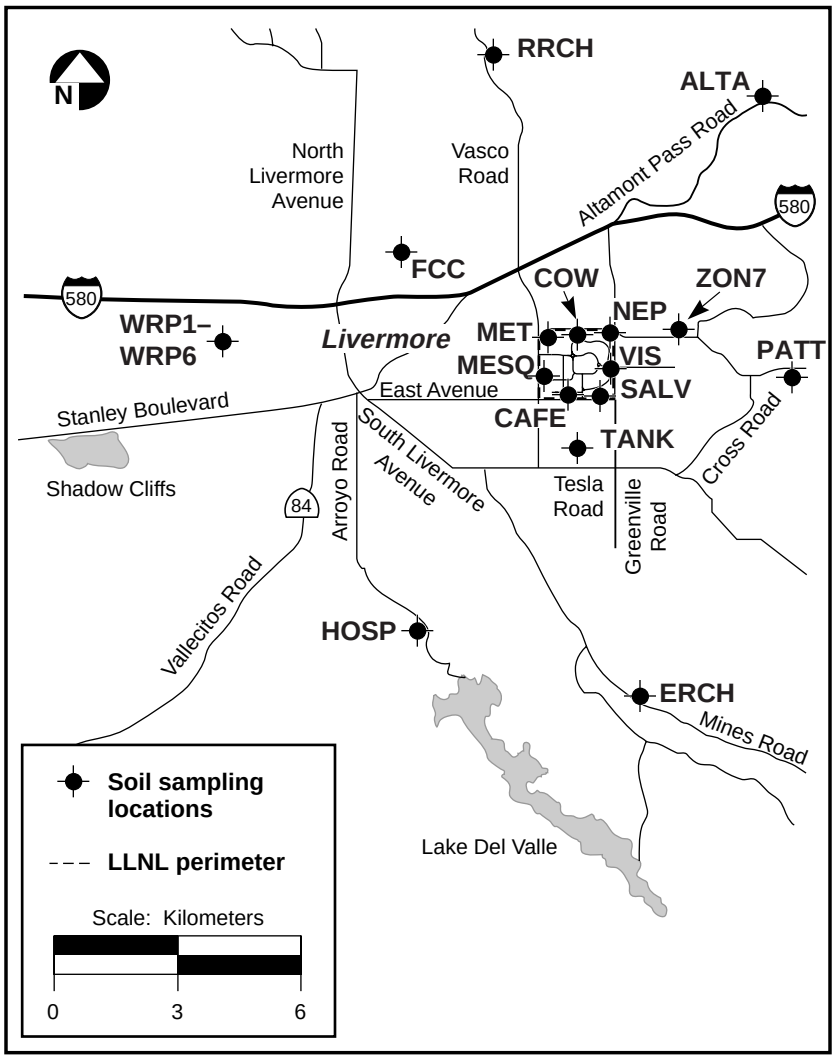

Figure 9-1. Soil sampling locations, Livermore Valley, 1996.

Soils and sediment samples are delivered to LLNL's Chemistry and Materials Science Environmental Services (CES) laboratory for analyses. Soil samples are dried, ground, sieved, and blended. The plutonium content of a sample aliquot is determined by alpha spectroscopy (Hall and Edwards 1994a, 1994b, and 1994c). Other sample aliquots (300 g) are analyzed for more than 150 radionuclides by gamma spectroscopy, using a high-purity germanium (HPGe) detector (Hall and Edwards 1994a, 1994b, and 1994c). The 10-g subsamples for beryllium analyses are sent to a contract analytical laboratory and are analyzed by graphite-furnace atomic absorption spectroscopy. For sediment samples collected for tritium analyses, CES uses freeze-drying techniques to recover water from the samples and determines the tritium content of the water by liquid-scintillation counting. Chain-of-custody procedures are followed throughout the sampling, delivery, and analytical processes.

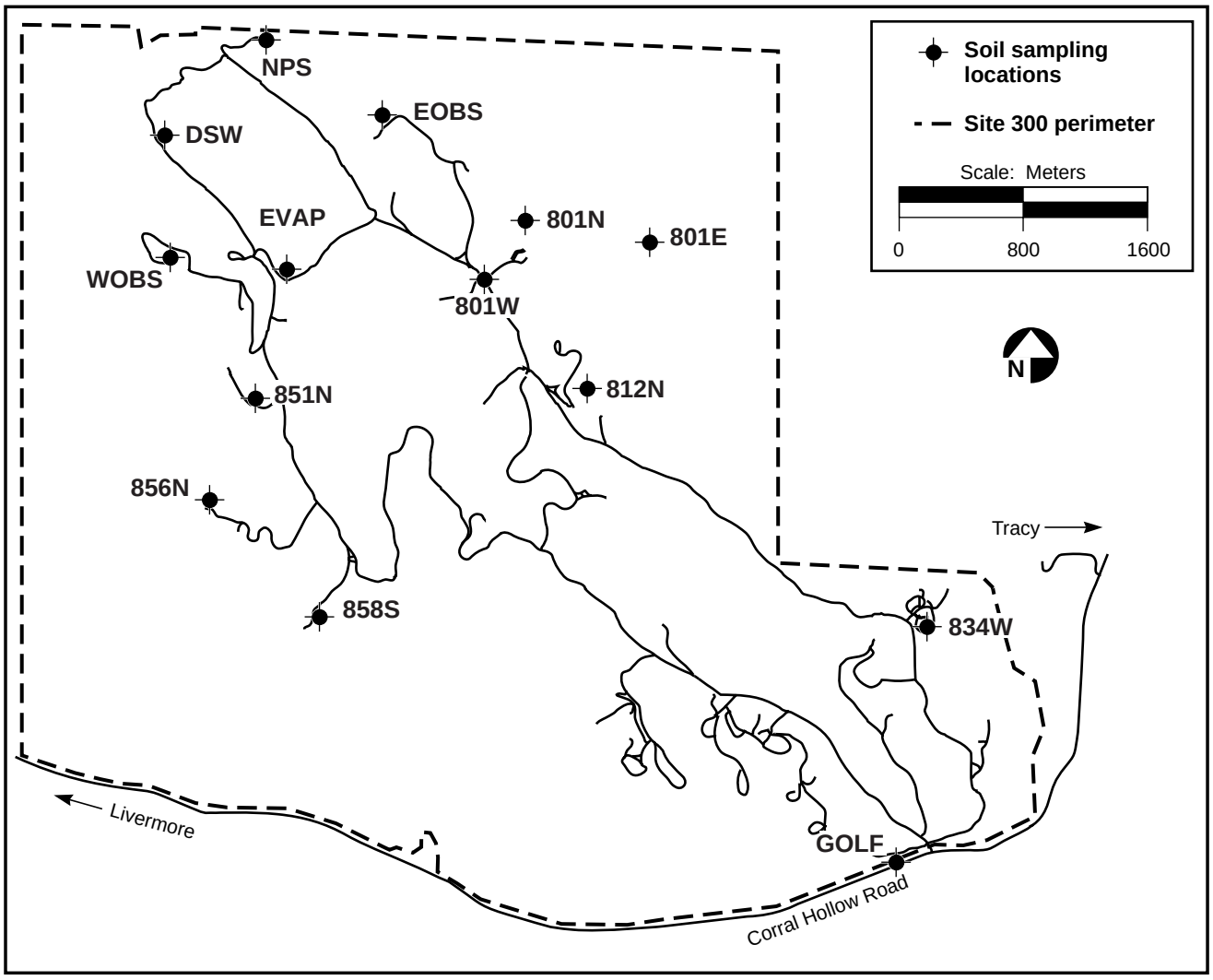

Figure 9-2. Site 300 soil sampling locations, 1996.

Livermore Valley Results

Table 9-1 presents summary data on the concentrations of $^{239+240}Pu$, ^{40}K, ^{60}Co, ^{137}Cs, ^{232}Th, ^{235}U, and ^{238}U in surface soils from the Livermore Valley sampling locations. The complete data for 1996 soils and sediment sampling is presented in Table 9-1, Volume 2, of this report. The concentrations and distributions of all observed

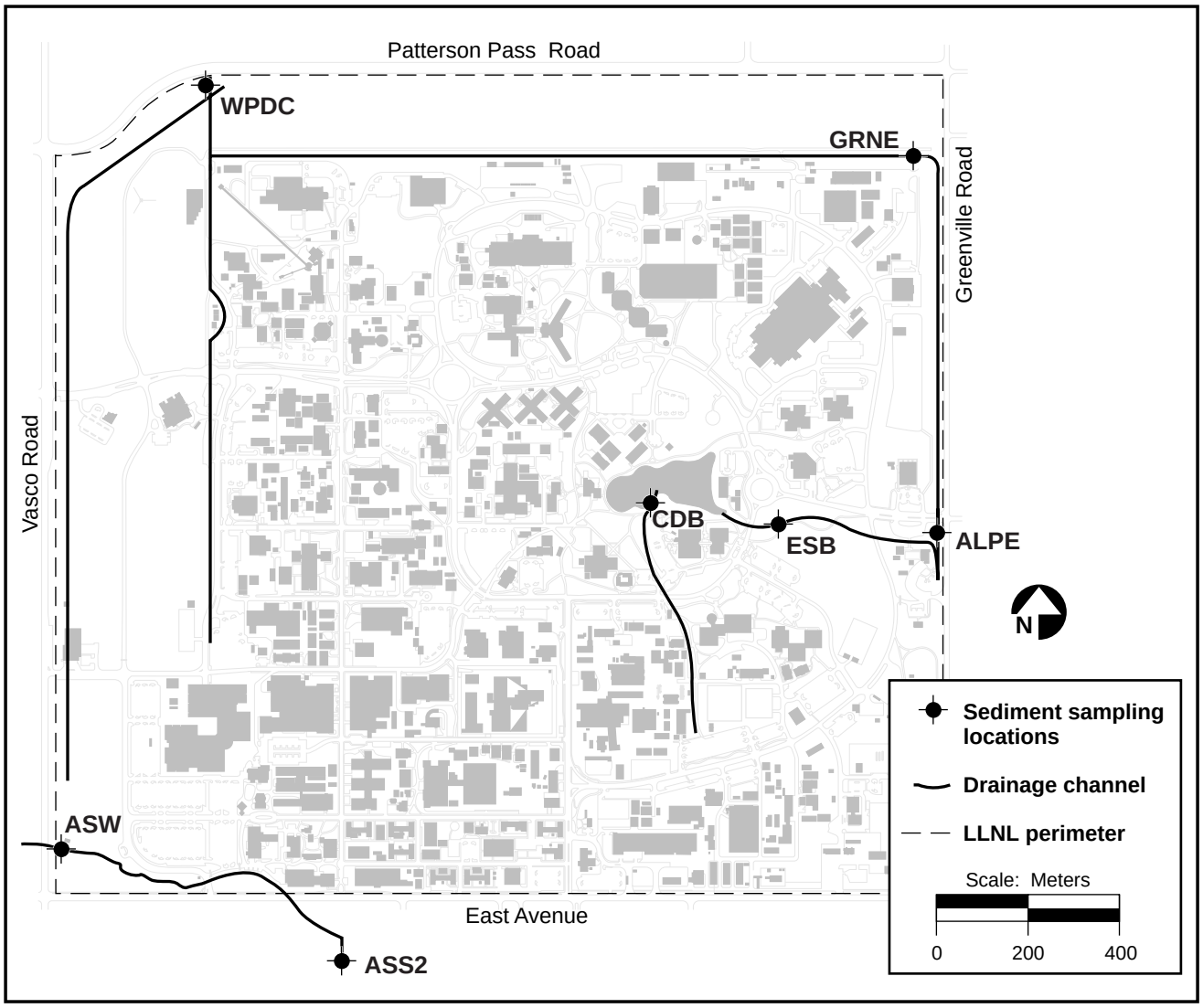

Figure 9-3. Arroyo and drainage basin sediment sampling locations, 1996.

radionuclides in soil for 1996 are within the ranges reported in previous years and generally reflect worldwide fallout and naturally occurring concentrations. The ratio of ^{235}U to ^{238}U generally reflects the natural ratio of 0.7%; however, there is uncertainty in the $^{235}U/^{238}U$ ratio because of the difficulty in measuring small quantities of ^{238}U by gamma spectroscopy.

Table 9-1. Summary of soils and sediment analytical data, 1996.

Analyte and location	Detection frequency[a]	Median	IQR[b]	Maximum
$^{239+240}$Pu (10^{-3} Bq/dry g)				
Livermore Valley soils	15/15	0.11	0.25	0.61
LWRP[c] soils	6/6	3.5	8.0	24
Livermore site sediments	6/6	0.029	1.3	2.2
Site 300 soils	14/14	0.079	0.064	0.20
^{137}Cs (10^{-3} Bq/dry g)				
Livermore Valley soils	15/15	3.1	1.7	7.1
LWRP soils	6/6	2.8	2.9	8.2
Livermore site sediments	5/6	0.40	0.42	0.94
Site 300 soils	14/14	2.4	1.9	6.4
^{40}K (Bq/dry g)				
Livermore Valley soils	15/15	0.470	0.124	0.585
LWRP soils	6/6	0.437	0.031	0.503
Livermore site sediments	6/6	0.509	0.100	0.577
Site 300 soils	14/14	0.477	0.089	0.622
^{232}Th (μg/dry g)[d]				
Livermore Valley soils	15/15	6.2	1.1	8.6
LWRP soils	6/6	6.8	0.6	7.5
Livermore site sediments	6/6	5.9	1.6	7.6
Site 300 soils	14/14	8.7	1.5	11
^{235}U (μg/dry g)[e]				
Livermore Valley soils	9/15	<0.022	—[f]	0.042
LWRP soils	6/6	0.025	0.002	0.029
Livermore site sediments	6/6	0.021	0.006	0.026
Site 300 soils[g]	13/18	0.030	—[f]	1.8
^{238}U (μg/dry g)[h]				
Livermore Valley soils	9/15	1.9	—[f]	3.8
LWRP soils	5/6	2.0	0.4	2.3
Livermore site sediments	3/6	<1.3	—[f]	1.7
Site 300 soils[g]	14/18	3.6	6.1	840

Table 9-1. Summary of soils and sediment analytical data, 1996 (concluded).

Analyte and location	Detection frequency[a]	Median	IQR[b]	Maximum
^{3}H (Bq/L extracted water)[i]				
Livermore site sediments	5/6	9.5	27	100
^{241}Am (10^{-3} Bq/dry g)[j]				
LWRP soils	3/6	<2.6	—[f]	5.1
^{60}Co (10^{-3} Bq/dry g)[j]				
LWRP soils	1/6	<0.072	—[f]	0.25
Be (mg/kg)[k]				
Site 300 soils[f]	16/18	1.3	0.7	53

a Detection frequency is the number of samples with results above the detection limit/the number of samples.

b IQR = interquartile range.

c LWRP = Livermore Water Reclamation Plant.

d Thorium-232 activities in Bq/dry g can be determined by dividing the weight in μg/dry g by 247.3, and pCi/dry g can be determined by dividing by 9.15.

e Uranium-235 activities in Bq/dry g can be determined by dividing the weight in μg/dry g by 12.5, and pCi/dry g can be determined by dividing by 0.463.

f Insufficient number of detections to calculate IQR. (See Site 300 results for discussion.)

g Includes results from reanalysis of original sample and analysis of resample.

h Uranium-238 activities in Bq/dry g can be determined by dividing the weight in μg/dry g by 80.3, and pCi/dry g can be determined by dividing by 2.97.

i Tritium (^{3}H) analysis is only conducted on sediment samples.

j Cobalt-60 and Americium-241 are only detected in LWRP soil samples.

k Beryllium analysis is only conducted on soils sampled at Site 300; the analysis is a chemical, not a radiochemical analysis.

Plutonium has, in the past, been detected at levels above background at ZON7, the off-site soils sampling location near the LLNL site and in the prevailing downwind direction. Because of the high level of variability inherent in the measurement of soils, we do not always find plutonium above background levels at this location. As in 1991, 1994, and 1995, $^{239+240}$Pu was detected at background levels—0.29×10^{-3} Bq/g (7.8×10^{-3} pCi/g)—at location ZON7 in 1996. Since 1973, soil samples in this area have generally shown $^{239+240}$Pu values that are higher than background. The slightly higher values near the Livermore site have been attributed to historic operations, which included the operation of solar evaporators for plutonium-containing liquid waste in the southeast quadrant (Silver et al. 1974). LLNL no longer operates the solar evaporators or engages in any other open-air treatment of plutonium-containing waste. Nonetheless, $^{239+240}$Pu from historic operations is carried off site by resuspension of soil by wind.

Similarly, elevated levels of $^{239+240}$Pu, resulting from an estimated 1.2×10^9 Bq (32 mCi) plutonium release to the sewer in 1967 and first observed in soils near LWRP during the early 1970s, again were detected at LWRP sampling locations. As in 1990 through 1992 and 1995, ^{241}Am was detected in LWRP samples; it is most likely caused by the natural decay of the trace concentrations of ^{241}Pu that were present in the release.

Historical plots of average $^{239+240}$Pu concentrations in soil in the Livermore Valley, at Site 300, and at LWRP are shown in **Figure 9-4**. Livermore Valley and Site 300 concentrations have remained relatively constant over the past 10 years and generally are indicative of worldwide fallout (locations on site and ZON7 show activities greater than background). Greater variability in $^{239+240}$Pu is seen at LWRP. However, six samples are evaluated to determine the median at LWRP. Moreover, the $^{239+240}$Pu is likely to be present in discrete particles, so the random presence or absence of the particles will dominate the measured $^{239+240}$Pu in any given sample.

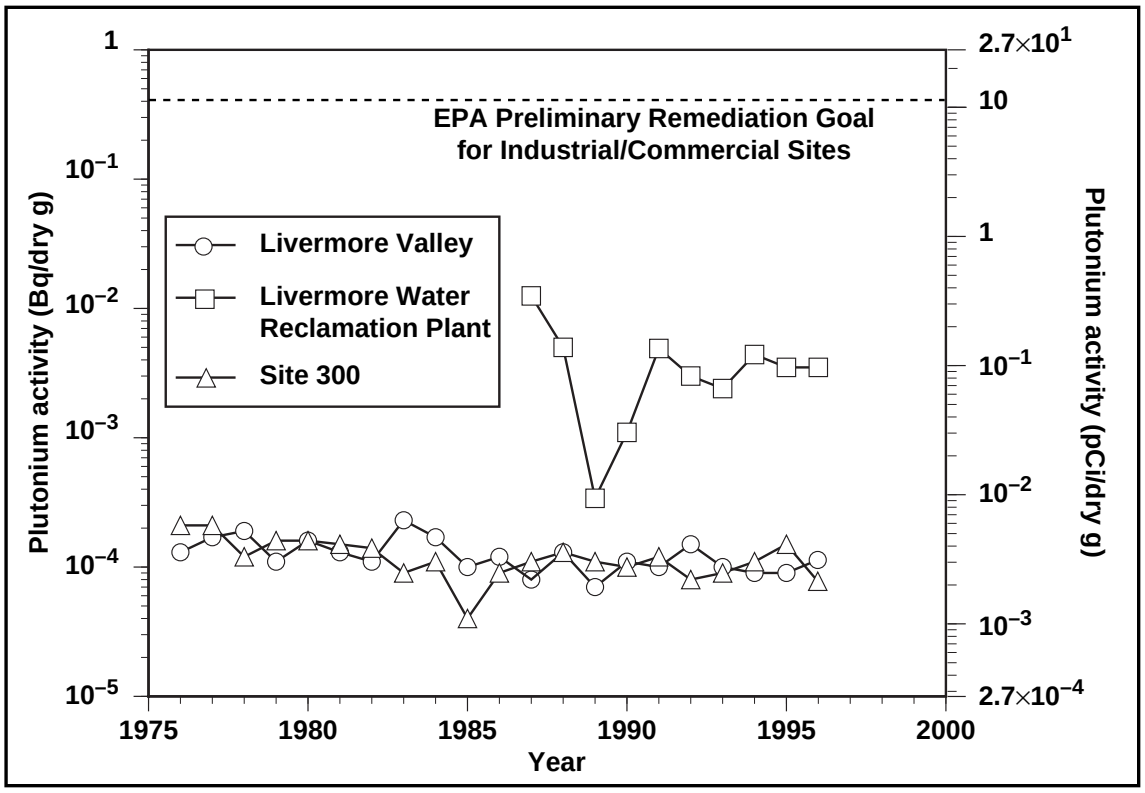

Figure 9-4. Median $^{239+240}$Pu activities in surface soils, 1976 to 1996.

Low levels of ^{60}Co were detected at the LWRP. While ^{60}Co is in use at the Livermore site, it is only present in gram quantities in two facilities (Buildings 151 and 514) or in sealed sources. Low levels of ^{60}Co, on the order of 0.0037 Bq / g (0.1 pCi / g), have also been detected intermittently in sewage sludge samples. If the Livermore site were the source of ^{60}Co, this activity of ^{60}Co in the sludge would translate into about 1.5×10^{-6} Bq / mL (40×10^{-6} pCi / mL) in the effluent leaving the site, which is below the detection limits of current analytical methods. This level is also well below the DOE effluent limit of 0.925 Bq / mL (25 pCi / mL). The reader should note that LLNL is not the only contributor to the waste stream that arrives at the LWRP and that ^{60}Co is used in a variety of medical, technical, and research applications. It is not possible to determine if LLNL is the source of ^{60}Co at LWRP. However, it can be concluded that LLNL controls on the release of ^{60}Co are sufficient to ensure that LLNL activities do not adversely affect LWRP applications.

Beryllium analysis for Livermore Valley soils was discontinued in 1995. The few LLNL operations that use beryllium are high-efficiency particulate air (HEPA) filtered. In addition, sampling data to date have shown no evidence of beryllium contamination in the Livermore Valley (Tate et al. 1995). Should beryllium usage change, LLNL's environmental monitoring staff would reevaluate the need for beryllium monitoring in soils.

Table 9-1 presents summary data on radionuclides detected in the sediment samples; a complete presentation of 1996 sediment data is found in Table 9-1, Volume 2, of this report. The levels of $^{239+240}Pu$ were generally at background concentrations, reflective of worldwide fallout. The slightly higher values at CDB and ESB may be attributed to historic activities in the southeast quadrant at LLNL; these locations are both in drainages for that area. Most other radionuclides were detected at levels similar to those reported from 1988 through 1995: ^{137}Cs, a fission product, was found at worldwide background concentrations; and ^{40}K, ^{232}Th, ^{235}U, and ^{238}U—naturally occurring radionuclides—were detected at background concentrations. Tritium concentrations were within the range of previous data. The data show a slight increase, which can be explained by the increase in tritium emissions from the Tritium Facility (see Chapter 5, Air Effluent Monitoring). Median tritium values are shown in **Figure 9-5**. There is not a ready explanation for the low results obtained in 1993. The sediment sampling procedure was revised in that year so that samples for gamma analysis were collected at the surface (5 cm deep). However, the depth for taking samples for tritium analysis was retained at 15 cm. Moreover, since samples were taken at the usual time, low values cannot be attributed to evaporative losses due to sampling delays. Tritium in sediments will continue to be evaluated.

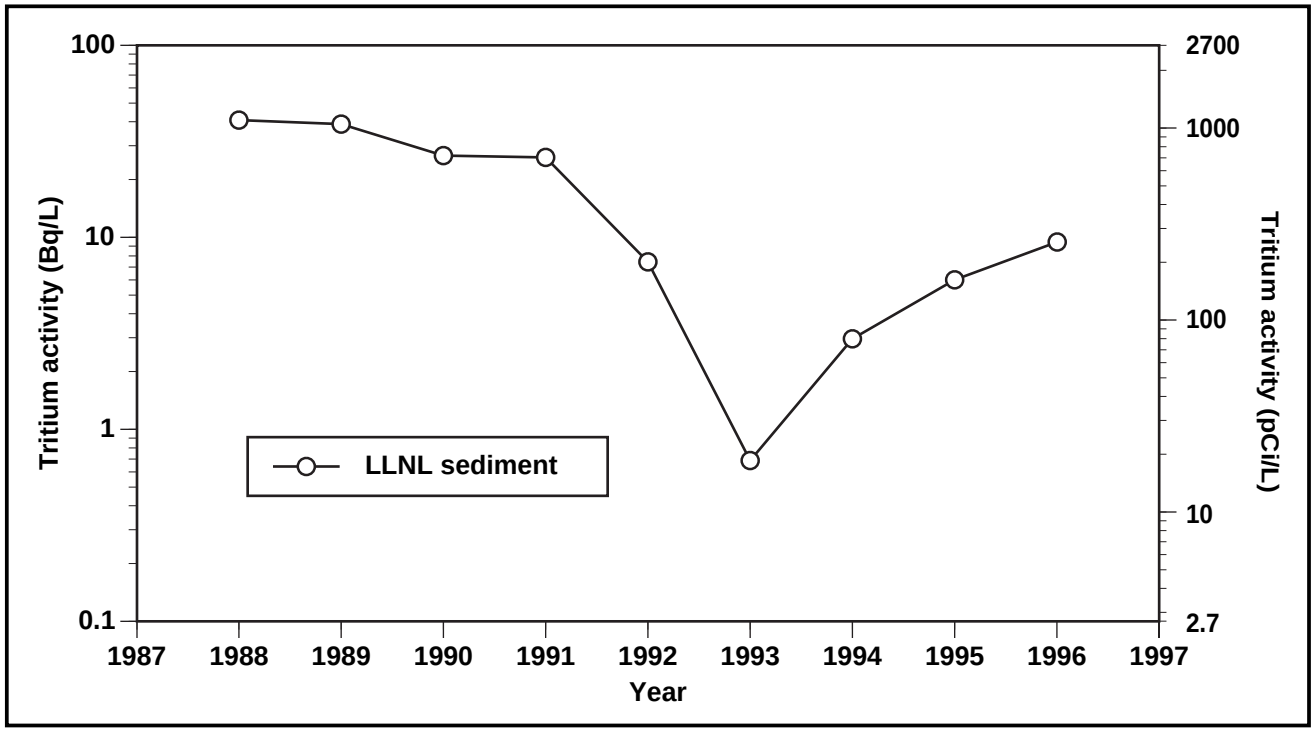

Figure 9-5. Median tritium concentrations in sediments (Bq/L of recovered water), 1988 to 1996.

Site 300 Results

Table 9-1 presents summary data on the concentrations of $^{239+240}$Pu, ^{40}K, ^{137}Cs, ^{232}Th, ^{235}U, and ^{238}U in soil from the Site 300 sampling locations; a complete presentation of 1996 soils data for Site 300 is found in Table 9-1, Volume 2, of this report. The concentrations and distributions of all observed radionuclides in Site 300 soil for 1996 lie within the ranges reported in all years since monitoring began, and, with the exceptions discussed below, reflect naturally occurring concentrations. The ratio of ^{235}U to ^{238}U generally reflects the natural ratio of 0.7%. Historical trends of ^{238}U concentrations from both the Livermore Valley and Site 300 are shown in **Figure 9-6**. Median values have remained relatively constant for both places. The highest values at Site 300 are caused by the use of depleted uranium in high-explosive tests.

During 1996, one sample from a region near a firing table (812N) had substantially higher than background concentrations of ^{238}U and beryllium. The ^{235}U/^{238}U ratios, at 0.2%, confirm the presence of depleted uranium; the ratio in naturally occurring material is 0.7%. To investigate the elevated ^{238}U and beryllium result at 812N, LLNL personnel

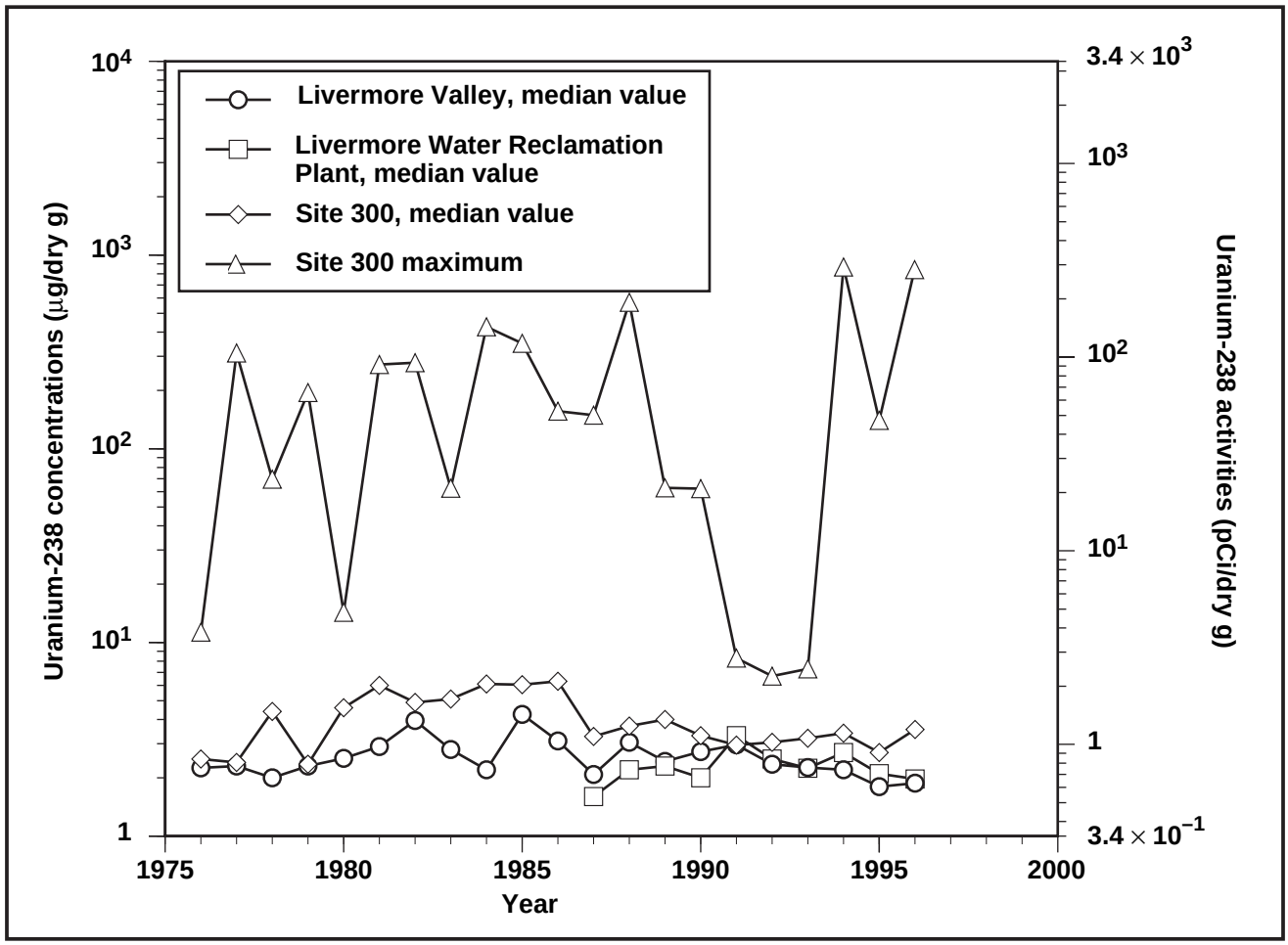

Figure 9-6. Uranium-238 activities in surface soils, 1976 to 1996.

resampled and analyzed the original sampling location. The high value of 32 μg/g of ^{238}U in the original sample was confirmed by reanalysis; the results of resampling were 35, 840, 6, and 140 μg/g. This disparity in sampling results was to be expected considering the heterogeneous nature of the contamination.

The finding of beryllium above background was also confirmed by resampling. The initial sampling yielded a result of 53 mg/kg. The results of resampling for beryllium were <0.5, 27, 7.7, and <0.5 mg/kg. Again, such heterogeneous results are expected because the contamination is not uniformly dispersed over the soil.

Environmental Impact

This section discusses the environmental impacts at the Livermore site and Site 300 inferred from soil and sediment monitoring.

Livermore Site

Routine soil and sediment sample analyses indicate that the impact of LLNL operations on these media in 1996 has not changed from previous years and remains insignificant. Most analytes of interest or concern were detected at background concentrations, in trace amounts, or could not be measured above detection limits.

The highest value of 24×10^{-3} Bq/g (0.65 pCi/g) for $^{239+240}$Pu measured at LWRP during 1996 represents 6.5% of the EPA preliminary remediation goal for commercial or industrial sites of 0.37 Bq/g (10 pCi/g) (U.S. EPA 1991). Statistical analysis shows that all LWRP $^{239+240}$Pu soils data are lognormally distributed, and at LWRP there is no general increase or decrease in $^{239+240}$Pu values with time. Moreover, all measured concentrations, regardless of location and year, have been a small fraction of the proposed EPA remediation goal, which is shown in **Figure 9-4** for comparison. Sampling of soils for radiological materials will continue on an annual basis.

Site 300

With the exception of elevated concentrations of ^{238}U and beryllium at location 812N, the concentrations of radionuclides and beryllium observed in soil samples collected at Site 300 are representative of background or naturally occurring levels. In 1988, contaminated gravel from the firing table at Building 812 was removed to on-site landfills, and measured values for samples from this location have generally not exhibited elevated levels of ^{238}U and beryllium. The elevated results for ^{238}U and beryllium indicate that areas outside the firing table may be contaminated by firing table debris. The investigation planned as part of the Site 300 CERCLA restoration efforts will clarify the nature and extent of the contamination in this area.